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# POLLUTED COASTAL WATERS: The Role of ACID RAIN

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## EXECUTIVE SUMMARY

Coastal waters of the eastern United States are receiving large inputs of nutrients, particularly nitrogen, which cause excessive growth of algae, loss of oxygen and light to the water and the long term decline of marine life. The major sources of nitrogen from human activity are thought to be runoff of fertilizer and animal waste from agricultural land, and the outfalls of sewage treatment plants and industrial facilities. The decline of the Chesapeake Bay and other estuaries is attributed in large measure to increasing nitrogen loadings, along with toxic chemicals, pesticides, and wetland destruction.

Here we report that atmospheric nitrate deposition, a component of acid rain, is a major source of nitrogen to the Chesapeake Bay and other Atlantic coastal waters. One-fourth of all nitrogen contributed by human activity to the Chesapeake Bay originates in acid rain and associated dry deposition falling directly on the Bay or onto its watershed. Atmospheric nitrate deposition exceeds sewage outfalls and runoff of animal waste as a nitrogen source to the Bay. Only fertilizer runoff surpasses atmospheric deposition as a source category, contributing one-third of the Bay's human-supplied nitrogen. Atmospheric nitrate deposition is a major factor in the decline of coastal waters.

In this study, the anthropogenic nitrogen source budget for the entire watershed was calculated, using data obtained from the Chesapeake Bay Program of the U.S. Environmental Protection Agency, the National Atmospheric Deposition Program, state agricultural agencies, and the U.S. Bureau of the Census. This source calculation was then refined to provide estimates of nitrogen input to the Bay itself from individual land use types using the limited available data on nitrogen retention. Previous studies have either ignored or seriously underestimated the contribution of atmospheric deposition to coastal waters.

Atmospheric nitrate deposited into this watershed arises from nitrogen oxide emissions due to fossil fuel combustion which are converted to nitrates in the atmosphere. The primary fossil fuel sources of nitrogen oxides are motor vehicles and electric power plants, which in 1984 contributed 36% and 32%, respectively, of the nitrogen oxides emitted in the eastern U.S. Nitrogen oxide emissions have increased more or less continuously since 1900, producing a steady increase in nitrate concentrations in surface waters across the eastern U.S. The problems of coastal waters discussed here are caused by the nutrient property of nitrogen, and are distinct from the problems of acidification arising in both sensitive fresh waters and some headwaters of the Chesapeake Bay.



Nitrogen oxide emissions are projected to increase another 40% to 60% over the next 40 years. Thus atmospheric deposition could become the major nitrogen source to the Bay over that period, particularly with control measures planned for other sources. In fact, any improvements in the nutrient status of the Bay due to reductions in other sources could be largely negated if nitrogen oxide emissions are not further reduced. Such reduction must include a Federal role since much of the nitrogen deposited inside the watershed arrives via long range transport from states that do not include parts of the watershed.

Atmospheric ammonium deposition contributes an additional 14% of the nitrogen reaching the Bay, so that the atmosphere is an important, and generally ignored medium for transfer of nitrogen in this watershed. Since ammonium arises largely from ammonia emissions related to agriculture, planned measures to control non-point sources may reduce this source contribution.

The Chesapeake Bay appears to be typical of other Atlantic coastal waters in regard to the role of atmospheric deposition. A preliminary analysis of nitrogen sources to Long Island Sound, and to the lower Neuse River, an estuary in North Carolina, indicates a similar source apportionment to the Chesapeake's, with nearly one-fourth of anthropogenic nitrogen inputs

originating in atmospheric nitrate deposition in each of these coastal waters.

The major recommendations of this report are:

- the federal government should adopt regulations for motor vehicles, power plants, and other stationary sources which result in a reduction of 40% in emissions of nitrogen oxides, consistent with reductions planned for other nitrogen sources to the Bay. Energy efficiency measures and alternative energy sources can contribute to the attainment of this goal, while reducing the other source of acid rain, sulfur dioxide emissions. These measures would provide benefits for other Atlantic coastal waters.
- states in the Chesapeake Bay watershed should implement stringent nitrogen oxide emissions limitations for new and existing stationary sources, while improving motor vehicle inspection and maintenance programs.
- both federal government and the states should vigorously pursue planned reductions in nitrogen from sewage treatment plants, as well as from fertilizer and other non-point-source runoff.

-----research programs should be undertaken to further refine our knowledge of nitrogen cycling in large watersheds. The contribution of atmospheric nitrate deposition to the nitrogen budgets of these watersheds should be examined in more detail.

Atmospheric nitrogen deposition is currently ignored in developing management plans for east coast waters. The implementation of the above recommendations would assure the gradual reduction of nitrogen inputs to these waters from fossil fuel emissions. In addition, it would result in decreased acidification of sensitive fresh waters, a decrease in air pollution from ground-level ozone (a major component of urban smog), and decreased nitrate in drinking water. If such measures are not implemented, the quality of east coast waters will continue to deteriorate.

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## I. Introduction

Over the past century, anthropogenic emissions of nitrogen oxides to the atmosphere of North America have exhibited steady increases.<sup>1</sup> These changes in the composition of the atmosphere have been reflected in increasing amounts of nitrate observed in precipitation and surface waters.<sup>2-4</sup>

Precipitation nitrate is an important, though secondary component of acid deposition, thereby contributing to the acidification of certain sensitive lakes and streams in the eastern U.S. and Canada<sup>5</sup>. In this report, we examine the contribution of atmospheric nitrate to a different problem, one which is pervasive along the eastern seaboard of the United States: the growth of nutrients in coastal waters, which has led to algal blooms, eutrophication and the destruction of marine life.

Studies have demonstrated that nitrate concentrations in fresh waters have increased on a regionwide basis over several decades<sup>2-4</sup>. Drainage of nitrate to north Atlantic coast estuaries, particularly the Chesapeake Bay, increased by about 30% over the period from 1974 to 1981<sup>3</sup>. Atmospheric deposition is an important external source of nitrate in some watersheds, contributing to the observed surface water nitrate

trends<sup>3,4</sup>.

Here we present the results of a study of the Chesapeake Bay watershed which demonstrates that atmospheric nitrate deposition is a major human induced source of nitrogen to the Bay's watershed. Indeed, its contribution approaches, or exceeds that of any other anthropogenic source of nitrogen, such as fertilizers, animal waste, and sewage and industrial waste water outfalls. Therefore, atmospheric nitrate is likely to be a leading cause of the eutrophication problem (cf. Section III), attributed in large measure to excess nitrogen, which has plagued the Chesapeake Bay. We examine briefly the nitrogen budgets of other Atlantic coastal waters and show that the Chesapeake appears to be typical of other estuarine waters in this respect. Some other areas where nutrient enrichment has occurred include the Long Island Sound, the New York Bight, Delaware Bay, and the Albemarle-Pamlico Sound, North Carolina.

Before examining the Chesapeake Bay and other coastal waters, we discuss the anthropogenic sources of nitrogen in surface waters and its ecological and health consequences. As an introduction to the problem of nitrate deposition, we summarize our previous analysis of external nitrate sources in an isolated watershed in the Catskill region of New York State. This study confirms that growth in atmospheric nitrate deposition has led to increases in surface water nitrate concentrations in waters which are affected in only small measure by other external sources.

We then move on to explore the more complex situation in the Chesapeake Bay watershed where other large anthropogenic sources contribute. Since nitrate is only one chemical form of the nutrient which is contributing to the Bay's eutrophication, we compare nitrate from precipitation to total nitrogen loading to the watershed from the other major anthropogenic sources.

## II. Anthropogenic Sources of Nitrogen in Surface Waters

Here we examine human influences on the nitrogen budget of surface waters. Major anthropogenic sources of nitrogen to surface waters are: agricultural runoff of both fertilizer and animal wastes, discharges from sewage treatment plants and industrial sources, and atmospheric deposition.

A. Commercial Fertilizer -- The use of commercial fertilizers has increased steadily in the U.S., adding nitrogen to agricultural soil.<sup>6</sup> Because most fertilizers contain a substantial proportion of nitrogen, and modern practice involves heavy applications to most farmland, runoff of agricultural fertilizer can be a large source of nitrogen compounds, especially nitrate and ammonium, to surface and ground waters. A national study of trends from 1974 to 1981 in inorganic nitrogen in U.S. rivers shows over eight times as many rivers experienced nitrate increases as experienced decreases in cropland drainages.<sup>2</sup> From 1975 to 1981 the total quantity of nitrogen fertilizer applied nationally increased by about 38 percent,<sup>7</sup> a change that contributed to the increasing nitrogen concentrations in surface waters of these watersheds.

In the Chesapeake Bay watershed, 17.9% of the land is cropland<sup>8</sup>. Although there has been a decrease of 23% in total harvested cropland in the basin from 1955 to 1980, (from 7.3 million acres to 5.6 million acres<sup>9</sup>), there was still a large



increase in total fertilizer nitrogen applied to the watershed. Between 1955 and 1970, fertilizer nitrogen application to the portions of Pennsylvania, Maryland, and Virginia in the watershed increased sharply, from 67,000 tons of nitrogen to 150,000 tons. Between 1970 and 1980, there was a much smaller increase, from 150,000 to 157,000 tons, but there is still an upward trend.<sup>9</sup>

B. Animal Waste-- Production of farm animals is an important industry in the watershed of the Chesapeake Bay. The inventory of animals on farms in this area includes over 2 million head of cattle, 1 million swine, 180,000 sheep and over 120 million chickens, turkeys, and other poultry. The waste produced by these animals is expected to be a significant contributor to nitrogen runoff in the Bay's watershed.<sup>10,11</sup>

C. Discharge from Point Sources-- Discharges of municipal and industrial waste waters, known as point sources, are concentrated sources of nitrogen released directly into surface waters. In the watershed of the Chesapeake Bay, there are over 6,000 point sources of nitrogen, which in 1985 discharged 27.9 million kg of nitrogen directly into the Bay, and another 13.0 million kg into rivers which drain into the Bay<sup>12</sup>.

D. Atmospheric Deposition -- Atmospheric sources generally have been considered minor contributors of nitrogen



compounds to surface waters. Due to increasing emissions of nitrogen oxides to the atmosphere over the last three decades, however, the amount of nitrogen reaching waters from precipitation may now equal or exceed inputs from other source categories.

Fossil fuel combustion is by far the major source of nitrogen oxide emissions in the U.S. Highway vehicles contributed 36% of nitrogen oxides to the atmosphere in 1984<sup>13</sup>, electric power plants contributed 32% and industrial combustion contributed another 15%<sup>13</sup>. According to 1980 data, other transportation activity contributed another 6 to 10%<sup>14</sup>. Natural sources of nitrogen oxide emissions in the U.S. are negligible in comparison.

Nitrogen oxides from combustion are eventually returned to the earth's surface in their gaseous form or as particulate nitrate, nitric acid or other soluble nitrates in wet precipitation or dry deposition. In certain oligotrophic waters nitrate loading from atmospheric sources may be the predominant form of nitrogen loading (cf Section V).

Ammonia from natural and agricultural sources is also an important source of atmospheric nitrogen deposition. In the Chesapeake Bay watershed, 36% of nitrogen in wet precipitation occurs as the ammonium ion arising from ammonia emissions (cf Section VI). A study of anthropogenic sources of ammonia emissions in Europe found that over 80% derived from decomposition of animal wastes, another 17% from volatilization

of fertilizer, and the rest from industrial sources<sup>15</sup>. Although the source apportionment may be somewhat different in the Chesapeake Bay region, we expect the sources of precipitation ammonium to be primarily agricultural, with a large fraction deriving from animal waste.

Nitrogen oxide emissions in the U.S. have increased substantially over recent decades.<sup>14,16,17</sup> From 1940 to 1980 nitrogen oxide emissions tripled,<sup>17</sup> although since 1978 this rate of escalation has declined. Nonetheless, in 1980 the United States emitted 21.3 million tons of nitrogen oxides into the atmosphere.<sup>1</sup>

Increasing nitrogen oxide emissions have been reflected in increasing nitric acid and nitrate concentrations in bulk precipitation at long-term United States Geological Survey (USGS) monitors, and at the Hubbard Brook Experimental Forest. Results of regression analyses for nine USGS precipitation stations in New York state indicate that nitrate increased at eight of these stations between 1965 and 1978<sup>18</sup>. Changes of analytical techniques during this period, however, limit the inferences that can be drawn from these regressions. Fifteen years of precipitation data collected at the Hubbard Brook Experimental Forest show significant increases in nitrate in precipitation. Nitric acid has also increased its contribution to the acidity of rainfall at Hubbard Brook<sup>5</sup>.

### III. Ecological Effects of Nitrogen

The ecological effects of nitrogen stem from its role as an important nutrient for both aquatic and terrestrial life. Nitrogen can cause eutrophication of certain aquatic systems<sup>19,20</sup>, as follows. Excessive nitrogen inputs can cause explosive growth of algae. These blooms of phytoplankton and benthic algae can be noxious to humans and directly toxic to aquatic animals, both fish and invertebrates. In addition, algal respiration and the decomposition of algal cells can rapidly deplete oxygen concentrations in a water body, and cause extensive kills of aquatic organisms. Algal blooms also increase the turbidity of the water, thus decreasing light levels to rooted aquatic plants. The significant decline in rooted plants in the Chesapeake Bay has likely resulted from decreases in light levels due to increased turbidity and growth of benthic algae on leaf blades<sup>21</sup>. Estuarine coastal waters, in which nitrogen is often a limiting nutrient<sup>19,22-24</sup>, are particularly vulnerable to this process.

Nitrogen in the form of atmospheric nitrate deposition also contributes to the problem of acidification of sensitive lakes and streams. As much as 40% of the acidity of "acid rain" in the Eastern U.S. is due to nitric acid. Over 3,000 lakes in the U.S., and another 10,000 in Canada have already been measurably acidified by acid rain.<sup>25</sup> Acid rain also causes billions of dollars per year in damages to building materials<sup>26</sup>, and is a

suspected contributor to in the current decline of forests observed throughout the eastern U.S.

The potential affect of the acidity of acid rain on the anadromous fish species (fish which travel upriver from salt to fresh water in order to breed, e.g. salmon) in Atlantic estuaries, was examined in a recent review.<sup>27</sup> The conclusion was that acidification of the freshwater rivers in which these fish breed is a possible contributing factor to the decline of these fish. Acidification of breeding areas was found to be particularly important in the decline of Atlantic salmon in the Southern Upland region of Nova Scotia<sup>27</sup>. Discharges of toxic metals and organic compounds, eutrophication, and overfishing are also thought to be contributing factors to the decline of Atlantic anadromous fish<sup>27</sup>. Although this is an important finding, we would like to emphasize that the question we are examining here is not the effect of rainfall acidity on estuaries, but rather the effect of rainfall nitrogen, and its contribution to eutrophication problems in estuaries.



#### IV. Health Problems Associated with Nitrate

Nitrate pollution of drinking water may cause health problems for humans, particularly in areas with intense agricultural activity. For instance, a study of 19 Illinois rivers by the Illinois State Water Survey from 1945-1970 found that after 1956 the nitrate health standard of 10 mg/l was equalled or exceeded in nine rivers, seven of which were used for drinking.<sup>6</sup> Although a detailed budget was not undertaken, agricultural runoff was considered the major cause of the upward concentration trend.

The known human-health effects of ingested nitrate occur after nitrate has been reduced to nitrite by microflora which subsist in the adult gastrointestinal tract where the pH is above 4. Nitrates are absorbed high in the gastrointestinal tract where the pH is usually below 4. However, in infants the pH of the upper tract is often above 4; consequently microflora migrate higher up the tract where they reduce nitrate to nitrite before the nitrate can be completely absorbed. Infants are particularly susceptible, therefore, to the health effects of ingesting nitrate.<sup>28</sup>

Nitrite can oxidize the iron of hemoglobin and render it ineffective as an oxygen carrier, resulting in asphyxiation and potentially death. This condition is known as infantile methemoglobinemia. No cases of infantile methemoglobinemia have been reported from the consumption of waters with less than 10

mg/l nitrate-nitrogen; hence the maximum contaminant level of nitrate in drinking water is set at 10 mg/l.<sup>28</sup>

Nitrite has other human health implications. Nitrite can combine with amines to form N-nitroso compounds, which have been shown to be carcinogenic, mutagenic and teratogenic in a wide range of animal species.<sup>28</sup>



## V. Atmospheric Deposition: a Controlling Factor for Surface Water Nitrate

The following is a summary of a previously published EDF analysis <sup>4</sup> which demonstrates that atmospheric deposition of nitrate can control surface water nitrate concentration at least for isolated watersheds. It was this study that led us to consider that atmospheric deposition might be an important contributor to surface water nitrogen concentrations throughout the eastern U.S. and Canada, even in bodies of water where there are other large, anthropogenic inputs of nitrogen.

An EDF study of 23 years of water quality data (1960 to 1983) from the Catskill reservoirs in upstate New York found that nitrate concentration had increased significantly at all four long-term measurement sites.<sup>4</sup> An average fourfold increase occurred in annual average values, with one site exhibiting a sixfold increase in nitrate levels over the period. Since historical data are available in this case for surface water concentrations and since the watershed budget is easily analyzed, the controlling effects of deposition are clearly demonstrable.

In determining the cause of the increasing nitrate trend in the Catskill reservoirs, we examined four possibilities: increases in the total amount of sewage input for the years 1960-1983, increased farmland runoff in the watershed, decreases in the rate of nitrogen uptake by the forested watershed, and

increased atmospheric deposition of nitrate.

An upper limit estimate of nitrate from sewage inputs showed that this source could account for no more than 17% of the total nitrate discharge from the watershed to the reservoirs. This contribution is too low for increased sewage discharge to be the cause of the observed increasing nitrate trend in the reservoirs. In addition, analyses of surface water ammonium for Catskill sites show no trends except in one case, where ammonium decreased. If sewage discharge were causing the increasing nitrate trend, it should also be causing an increasing ammonium trend because ammonium is the predominant form of nitrogen in wastewater<sup>29</sup>.

Data from the Census of Agriculture for the 2 counties which contain the watersheds (Ulster and Greene) <sup>30</sup> show that during the period from 1964-1982 these two counties lost 44% of their farmland. Data in state agricultural reports also showed that fertilizer applied to Ulster county was less in 1982 than 1969, and only slightly higher in Greene county. Based on these observations farmland runoff is precluded as a cause of the increasing surface water nitrate trend. The lack of a coincident trend for ammonium also precludes farmland runoff, since ammonia is a major component of most modern fertilizers and is also emitted by animal waste. Ammonium also should be found in higher concentrations in surface water if agricultural sources of nitrate increase<sup>29</sup>.

Decreased nitrate retention by the forested ecosystem of

the Catskill reservoirs might explain the increased nitrate levels in the reservoirs, particularly if there had been a large loss of forested land over the period. However, given the large loss of farmland in the region, it is likely that the amount of forested land has increased. Given this land use shift, the nitrate trend appears to be both too large (fourfold increase), and too monotonic to be explained by a change in nutrient cycling.

Atmospheric deposition from nitrogen oxide emissions can be a major source of nitrate into an ecosystem. According to our calculations, sufficient nitrate has been deposited from the atmosphere to account for the nitrate observed in the Catskill reservoirs.

The trend toward increasing nitrate concentrations noted in Catskill reservoirs has been observed for many water bodies in the Eastern U.S., indicating a regional source<sup>2</sup> (See Appendix I for a more detailed discussion of this trend). The eastern half of the country as well as the entire U.S has experienced increasing nitrogen oxides emissions, which correspond roughly with surface water nitrate concentration trends in the Catskill reservoirs and elsewhere<sup>4</sup>. Over the same period, nitrate concentrations at the nearby Rock Hill, N.Y., USGS bulk precipitation monitor increased steadily<sup>18</sup>. Similar trends are observed for Adirondack surface waters<sup>31</sup>. We concluded that increases in nitrogen oxide emissions caused the changes of nitrate concentrations in the reservoirs. No other source can

adequately explain the observed trend.

The analysis demonstrated that atmospheric deposition is the probable cause of long term increases in nitrate concentrations in a relatively isolated watershed. This trend is a concern because of the health effects of increasing nitrate in this drinking water supply, and because nitric acid can contribute to acidification. However, since freshwater productivity is generally limited by phosphorus, which is extremely low in the Catskill reservoirs, eutrophication is not a problem in this system.

In the following analysis we examine a more complex system, the Chesapeake Bay, which has larger external sources of nitrogen from sewage and agriculture, to determine if atmospheric deposition still plays an important role in the nitrogen budget. Here the issue is of great ecological importance since eutrophication is already a major, widespread concern, and reduced nitrogen loadings seem necessary to reduce this problem.

## VI. The Chesapeake Bay -

### A. Introduction

The Chesapeake Bay is one of the largest estuary systems in the world. It is 195 miles long, ranges from 3.4 to 35 miles wide, has an area of 4400 square miles (including tidal portions of its tributaries), and has 4600 miles of shoreline, 1750 of which are navigable. Although its average depth is only 28 feet, it has a deep natural channel running the length of the bay.<sup>32</sup> It also contains many man-made channels, including a major navigation channel constructed by the Army Corps of Engineers to Baltimore.

The drainage basin of the Bay (See Figure 1), 64,000 square miles in size, covers portions of six states and the District of Columbia. It includes over 150 rivers, creeks, and branches, 50 of which are considered major tributaries. The six largest of these contribute almost 90 percent of the freshwater input into the Bay. The largest single contributor is the Susquehanna River, accounting for 50% of the freshwater input to the Bay. The Susquehanna's 440 mile length and 27,500 square mile basin begins in the Finger Lakes region of New York, and extends through Pennsylvania to the head of the Bay in northeastern Maryland. The next largest tributary is the Potomac River, with a drainage basin of 14,670 square miles encompassing parts of four states and all of the District of Columbia. The other four



# Chesapeake Bay Watershed

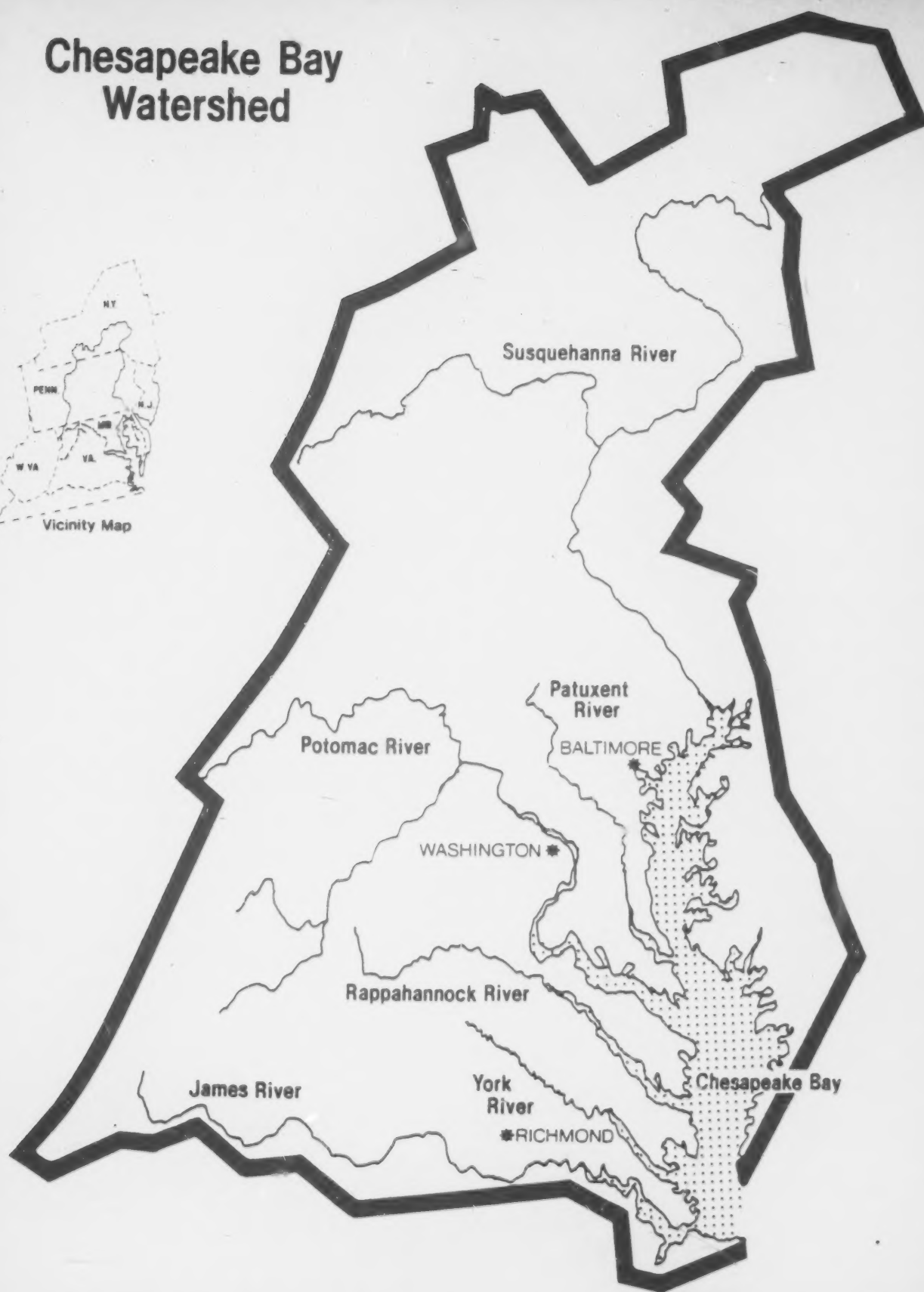
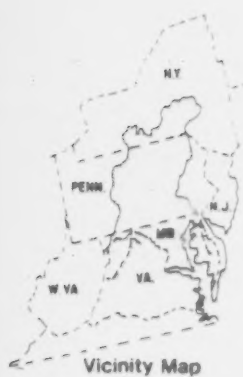


Figure 1

tributaries which complete the top six are the Rappahannock, the James, the York, and the Patuxent Rivers.<sup>33</sup>

The Chesapeake Bay is a valuable resource, whose preservation is vital to the economy as well as the ecology of the region. It is a productive source of oysters, blue crabs, menhaden, rockfish, and other seafood. An important habitat for wildlife, the Bay provides a stop along the Atlantic Flyway for migratory birds and waterfowl, and spawning grounds for many species of saltwater fish. The Bay contains two of this country's five major North Atlantic ports, Baltimore in the north Bay, and the Hampton Roads Complex in the south. Industries rely on the Bay as a source of water for industrial processes and cooling. Many of the 13 million inhabitants of the Bay's watershed rely on it for their economic livelihood. The Bay is also a unique recreational resource<sup>34</sup>.

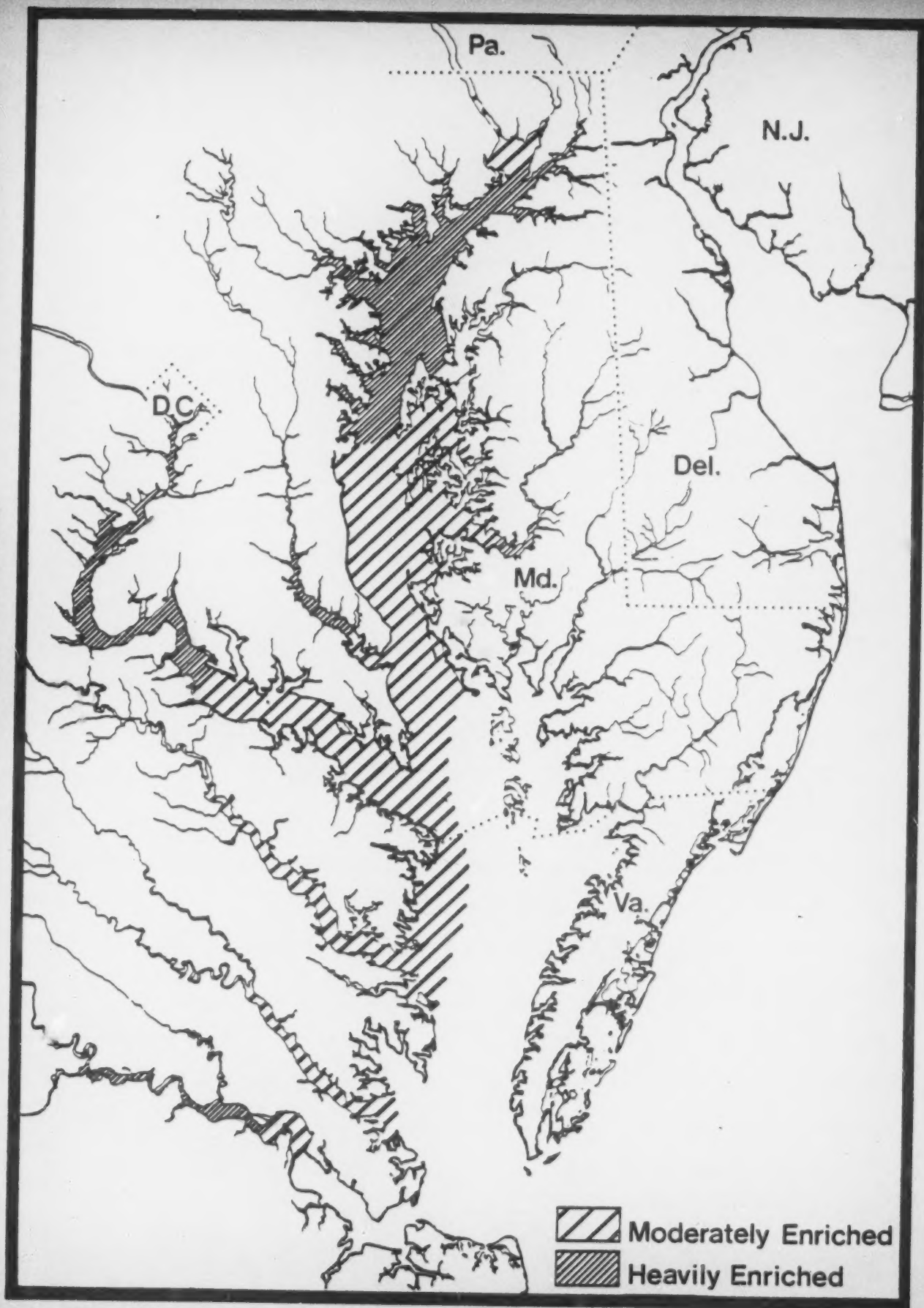
## B. Water Quality in the Bay<sup>35</sup>

Water quality has been deteriorating in the Chesapeake Bay since the time of the Industrial Revolution. This decline has accelerated in the last 30 years. One of the major causes of this decline is increased nutrient loading, which leads to the eutrophication process described in Section III. The main nutrients of concern are nitrogen and phosphorus. Nutrient enrichment (as defined by Heinle et al.<sup>36</sup>) is most severe in the upper reaches of the Chesapeake Bay, and is also problematic in the mid Bay region (See Figure 2, taken from Reference 36). It does not seem to be a major problem yet in the lower portions of the Bay.

Excessive nutrient loading in the Chesapeake Bay has led to eutrophication and a host of related problems. Massive blooms of harmful blue-green algae have been observed frequently in the upper Bay in recent years; in the 1960's such blooms occurred only occasionally. The number of algal cells in this area was estimated to have increased 250-fold between 1950 and 1980.

Algal blooms are associated with decreased levels of dissolved oxygen (DO), since algal metabolism and the decay of dead algal cells consumes oxygen. This problem is especially severe in benthic (bottom) regions, harming benthic organisms, including shellfish, and the organisms which feed on them. The volume of Bay water with less than 0.7 mg per liter DO increased 15-fold between 1950 and 1980. In the same time period, the

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Excess Nutrients in the Bay



area affected by low DO (0 to 3 mg per liter) increased from a few limited regions, to a major portion of the main stem, and the duration of low DO levels increased from a few weeks in the summer, to include the beginning of May through mid-September<sup>35</sup>. Algal blooms also lead to increased turbidity and consequent reduction of light levels reaching submerged vegetation.

Changes in the Bay which may be attributable in whole or in part to decreased DO and decreased light levels include:<sup>35</sup>

- decreases in amount and diversity of submerged aquatic vegetation (SAV)
- corresponding changes in the populations of waterfowl which feed on SAV
- decreased oyster harvests in the Upper Bay
- decreases in amount and diversity of other benthic organisms
- decreases in populations and diversity of species of fish which spawn in fresh water

There are other confounding factors which may also contribute to the above problems. These include increasing levels of toxic compounds, such as heavy metals, pesticides and herbicides in the Bay. These factors may exacerbate the detrimental effects of nutrient enrichment and consequent anoxia.



### C. Limiting Nutrient

There are several essential nutrients that all micro-organisms need in order to grow. Either nitrogen or phosphorus may become limiting in some circumstances. In cases where one nutrient is limiting, additional inputs of the other nutrients will not cause additional growth of the micro-organism.

The question of whether phytoplankton production in a eutrophied body of water is nitrogen or phosphorus limited has important policy implications. If algal growth is solely limited by one nutrient, then controlling the other will have little effect. Primary production is generally believed to be phosphorus limited in freshwater and nitrogen limited in marine waters <sup>23</sup>. In estuaries, the question of which nutrient is limiting at what time of year has been the subject of much study and controversy.

There are several techniques which scientists and managers have used to assess nutrient limitation status in estuaries. These include <sup>23</sup> measuring some metabolic characteristic of phytoplankton to indicate nutrient status, measurement of relative availability of inorganic nutrients, use of models to predict responses to nutrient loading, or "growth bioassays" (described below).

D'Elia <sup>19,23</sup>, Correll<sup>37</sup>, and others<sup>38-40</sup> have argued convincingly that the best way to determine if algae is nitrogen or phosphorus limited is to "ask the algae" by

conducting bioassays. In D'Elia's experiments, controlled amounts of nitrogen or phosphorus were added to large continuous cultures of phytoplankton in water from the lower Patuxent River (a sub-estuary of the Chesapeake Bay). Observations were made of the amount of growth induced by either phosphorus or nitrogen, compared to a control. It was found that nitrogen strongly stimulated growth during summer and fall (June through November), increasing observed biomass by a factor of anywhere from 3 to 14. Response to increased nitrogen was quite rapid, with biomass doubling in 1 to 2 days. Phosphorus stimulated growth in November through February, but a substantially smaller increase was observed, no more than a factor of 3. The response was also less rapid, taking as long as 10 to 15 days. The authors note that such a slow, weak phosphorus response does not offer much insight into the nutrient limitation status of the phytoplankton, since a response of this type could be an artifact of experimental conditions.<sup>23</sup> It is likely that the colder temperatures of winter retard the response of the algae.

The same summer to fall period during which phytoplankton growth tends to be nitrogen limited is also the period during which nuisance algal blooms occur, and anoxia becomes a problem. Increased nitrogen inputs at this time of year can be expected to increase the amount of algae.

Nitrogen inputs at other times, particularly late winter and spring, influence production of algae as well. It is at

this time that estuaries receive their largest flows of freshwater, and consequently their largest inputs of non-point source (NPS) nutrients, the major source of nitrogen input. In studies conducted in the estuarine portions of the lower Neuse River in North Carolina between 1981 and 1986, Paerl<sup>41</sup> observed that large inputs of nitrogen during late winter to spring, combined with low flow conditions during the summer were controlling factors in determining the severity of algal blooms occurring in the summer. Under summertime low flow conditions, the Neuse River behaves essentially like a lake or reservoir, with flow rates reduced to nearly zero. Residually high nitrogen, left over from the winter-spring period, becomes trapped due to the low flow, feeding the growth of summertime blooms. Based on his observations, Paerl recommends targeting nitrogen input cutbacks in this estuary as early as February.

How long the Chesapeake Bay system retains nutrients, especially nitrogen, is the subject of some controversy. However, estimates range from 6 months to a year at the low end<sup>42</sup>, (based on the flushing time of the Bay) to essentially infinite retention times at the high end<sup>43,44</sup>. Even if the shorter estimate is correct, it is clear that winter-spring inputs will persist long enough to influence summertime blooms.

From the above studies it appears that limitation of algae growth would be most effective with year-round control of nitrogen inputs. In order to develop management plans for controlling nitrogen, it is important to understand its sources.

In the next section we discuss the major sources of nitrogen to the Bay.

#### D. Results of Previous Studies

Previous studies identify the major sources of nitrogen into the Chesapeake Bay as agricultural runoff from both fertilizers and animal wastes, and effluent from sewage treatment plants (STP's) and industrial sources. STP's and industrial sources are point sources which discharge directly into surface waters. The other sources are non-point sources (NPS).

There are only two studies we are aware of which look at the contribution of atmospheric deposition to nitrogen budgets in the Chesapeake Bay or a subestuary. In both of these studies, only the contribution of direct nitrogen deposition to the estuary itself was considered. The added contribution of atmospheric nitrogen deposition via runoff from the watershed was not included.

In a 1982 study,<sup>43</sup> Smullen et al. calculated the total nitrogen input to the Chesapeake Bay from atmospheric sources, rivers, point sources, and "benthic sources" (release of nitrogen stored in sediments). They found that direct input of atmospheric nitrogen to the Bay was 13.3% of the total nitrogen input. The "benthic source" of nitrogen is an internal nitrogen source. Of the external sources, direct atmospheric nitrogen deposition represents 14.8% of the total input. Five percent of the total input was nitrate nitrogen, but atmospheric deposition was measured by a method (open buckets to collect dry and wet



deposition) which is known to give only a lower bound on total nitrate deposition, because gaseous deposition is not efficiently measured. This factor is particularly important when deposition to the terrestrial watershed is considered.

A study conducted on the Rhode River, a subestuary of the Chesapeake Bay, compared nitrogen loading from direct deposition on the river with loading from land runoff. It found that loading from deposition exceeded that from land runoff during the summer, and that of the total annual average input of 12.4 tons of nitrogen, direct deposition on the surface of the river contributed 5.1 tons and watershed runoff contributed 7.3 tons, making the sources comparable. During the summer, nitrate nitrogen from direct deposition was almost double the nitrate nitrogen from land runoff, and annually deposition contributed 2.3 tons of nitrate nitrogen, compared to 3.0 tons from land runoff. This study also used the open bucket method of measuring direct deposition, thus nitrate loadings from deposition are lower bound estimates. Atmospheric nitrate deposition contributes a minimum of 19% of the total nitrogen input to this estuary. Nitrate in bulk precipitation at the Rhode River site increased steadily for the 7 years of the study, and the report concludes that, from a long-term perspective, nitrogen loading of the Rhode River Estuary via precipitation is very important, and provides 50 percent of the annual readily-available nitrogen inputs to the estuary<sup>45</sup>.

These two studies establish that the contribution of direct

atmospheric deposition of nitrate to the Bay is by itself an important source of nitrogen. However, such analyses underestimate the importance of this source, since atmospheric deposition to the watershed of the Bay will contribute further to nitrogen loadings via land runoff.

#### E. Nitrogen Loadings to the Chesapeake Bay's Watershed

In order to place some bounds on the relative importance of each nitrogen source category (deposition, fertilizers, and animal waste) to land runoff, we have calculated nitrogen loadings to the entire watershed from these sources. The calculations we performed are described in detail in Appendix II. Table I shows the result. Figure 3 shows the fraction of nitrogen loading to the Bay's watershed from each source category in 1984. We chose to work with 1984 data because this was the most recent year for which relatively complete data on all source categories were available.

We calculated wet deposition of nitrate and ammonium nitrogen to the watershed using monitoring data from the nearest National Acid Deposition Program (NADP) monitoring site for each sub-basin listed in Tables IIA-C. To calculate total nitrogen deposition, we then assume that dry deposition is approximately equal to wet deposition, and multiply the wet deposition total by a factor of two.

This calculation of total deposition should produce a conservative value, for two reasons. First, although dry deposition is difficult to measure accurately, several researchers have determined it for nitrogen by a variety of techniques.<sup>46-49</sup> In these studies, total nitrate deposition to land is found to exceed wet deposition by a factor of 2 to 5. The factor of 2 which we have used is at the low end of this

Table I  
CALCULATED NITROGEN LOADINGS TO CHESAPEAKE BAY WATERSHED, 1984

| Source        | 10 <sup>6</sup> kg/year | kg/ha-yr | % of total | % of NPS |
|---------------|-------------------------|----------|------------|----------|
| precipitation |                         |          |            |          |
| nitrate       | 143                     | 8.7      | 23%        | 25%      |
| ammonium      | 79                      | 4.8      | 13%        | 14%      |
| animal waste  | 195                     | 11.9     | 32%        | 34%      |
| fertilizer    | 158                     | 9.6      | 25%        | 27%      |
| NPS subtotal  | 575                     | 35.1     |            | 100%     |
| point sources | 41                      | 2.5      | 7%         | -        |
| total         | 616                     | 37.6     | 100%       | -        |

## Sources of Nitrogen—The Watershed

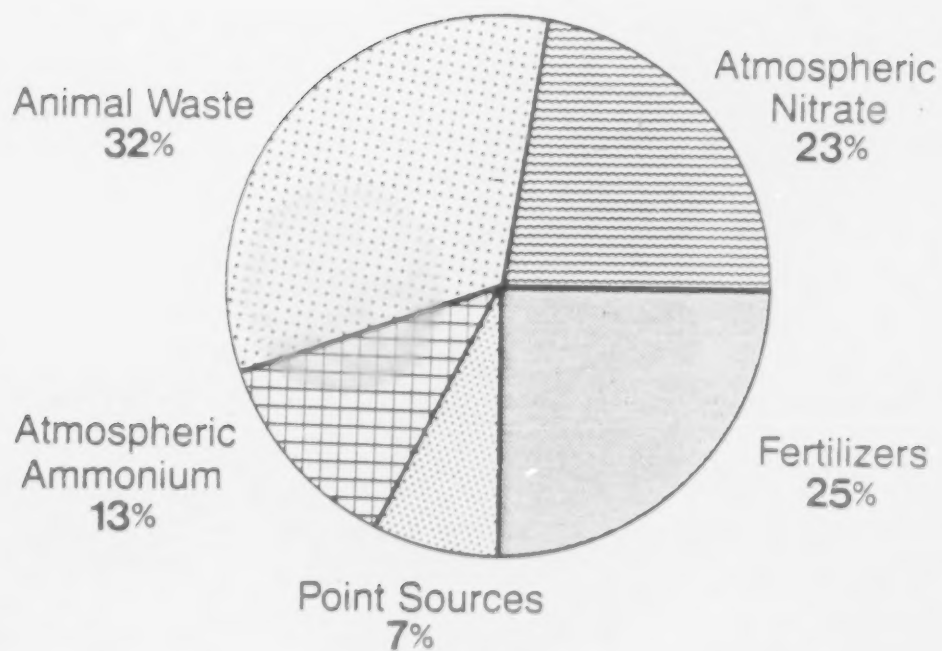


Figure 3



Table IIA  
DRAINAGE BASINS AND NADP MONITORS

| Basin             | Nearest NADP monitor          |
|-------------------|-------------------------------|
| Susquehanna       |                               |
| N. Branch         | Aurora, Cayuga county, NY     |
| W. Branch         | Penn State, Centre co., PA    |
| below Sunbury     | Leading Ridge, Huntington, PA |
| W. Chesapeake     | Wye, Queen Anne co., MD       |
| Patuxent          | "                             |
| Eastern Shore     | "                             |
| Potomac River     | "                             |
| York-Rappahannock | "                             |
| James River       | "                             |

Table II-B  
WET DEPOSITION RATES AT NADP MONITORS, 1984

| monitor       | nitrate nitrogen<br>(kg/ha-yr) | ammonium nitrogen<br>(kg/ha-yr) |
|---------------|--------------------------------|---------------------------------|
| Aurora        | 4.63                           | 3.11                            |
| Penn State    | 5.49                           | 2.72                            |
| Leading Ridge | 5.69                           | 2.80                            |
| Wye           | 3.66                           | 2.02                            |

Table II-C  
ATMOSPHERIC NITROGEN DEPOSITION, BY DRAINAGE BASIN

| Basin                                     | area<br>(hectares) | nitrate<br>(million kg nitrogen) | ammonium |
|-------------------------------------------|--------------------|----------------------------------|----------|
| Susquehanna                               |                    |                                  |          |
| N. Branch                                 | 2,905,000          | 13.45                            | 9.04     |
| W. Branch                                 | 1,834,000          | 10.07                            | 4.99     |
| below Sunbury                             | 2,327,000          | 13.24                            | 6.52     |
| W. Chesapeake                             | 415,000            | 1.52                             | 0.84     |
| Patuxent                                  | 244,000            | 0.89                             | 0.49     |
| Eastern Shore                             | 1,103,000          | 4.04                             | 2.23     |
| Potomac                                   | 3,653,000          | 13.36                            | 7.38     |
| York-Rappahannock                         | 1,445,000          | 5.29                             | 2.92     |
| James River                               | 2,559,000          | 9.37                             | 5.17     |
| total, wet deposition                     |                    | 71.26                            | 39.58    |
| total, including estimated dry deposition |                    | 142.52                           | 79.16    |

range. It is more difficult to estimate dry deposition of ammonia, but a factor of 2 is a reasonable first approximation if gaseous ammonia, rather than particulate ammonium sulfate, is the dominant source<sup>50</sup>. For both species, dry deposition to water surfaces is expected to be slower than to land, so our calculation may overestimate the importance of direct deposition of atmospheric nitrogen to the Bay itself, which is a minor portion of the total watershed area.

Second, our reliance on NADP data will slightly underestimate wet deposition. Nitrate deposition is known to be higher at higher elevations than at lower ones.<sup>51</sup> However, NADP stations are generally located at low to moderate elevations. Since the watershed contains both high and low elevations, use of these data will give us a low estimate of total deposition.

If our estimate of nitrogen loading to the watershed is reasonable, it should be considerably larger than estimates of total loadings of nitrogen to the Bay itself, since a significant fraction of the watershed loading is expected to be consumed by biological, chemical, and physical processes on land and in the rivers before it reaches the Bay. Table III summarizes the US EPA Chesapeake Bay Program's estimate of nitrogen loadings to the Bay.<sup>52</sup> The Chesapeake Bay Program is currently revising the model which generated these loadings, which incorporate assumptions about terrestrial and riverine retention of nutrients. However, the total loadings from the

Table III  
ESTIMATED NITROGEN LOADINGS TO CHESAPEAKE BAY\*

| At Fall Line    | dry year | average year | wet year |
|-----------------|----------|--------------|----------|
| point sources   | 5.1      | 6.3          | 7.8      |
| - non-point     | 41.6     | 50.6         | 103.6    |
| sub-total       | 46.7     | 56.9         | 111.4    |
| Below Fall Line |          |              |          |
| point sources   | 26.6     | 26.6         | 26.6     |
| non-point       | 10.7     | 16.2         | 41.5     |
| subtotal        | 37.3     | 42.8         | 68.1     |
| Watershed total |          |              |          |
| point sources   | 31.7     | 32.9         | 34.4     |
| non-point       | 52.3     | 66.8         | 145.1    |
| TOTAL           | 84.0     | 99.7         | 179.5    |

\* from US EPA (1983), "Chesapeake Bay: A Framework for Action". This publication reports loadings for March through October (8 months). We approximate annual loadings by multiplying by a factor of 1.5. This should be a good approximation for loadings due to point sources, but may overestimate loading resulting from fertilizer application, since this is mostly applied in the spring.

model agree well with measurements of nitrogen loading to the Bay via river inputs <sup>43,53</sup> plus point source inputs below the Fall Line (the line below which land runoff and point sources are discharged directly into the Bay), so the total loading is not expected to be revised significantly. Since most of the point source loading comes in below the Fall Line this factor is also only weakly dependent on the model assumptions about riverine uptake, so the contribution from point sources is not expected to change much either.

Our calculated loadings to the watershed are larger by a factor of 3.4 to 7.3 than estimated loadings to the Bay itself (wet vs. dry year). Our estimate of NPS source loading to the watershed is 4.0 to 11.0 larger than EPA estimated non-point source loading to the Bay. This means that our estimate of NPS nitrogen loading to the watershed is reasonable if 75% to 91% of the NPS nitrogen is consumed by watershed processes before reaching the Bay. This range is consistent with the values observed in small watershed studies<sup>4,11,46,54-58</sup>.

It is clear from Table I that atmospheric deposition is a significant source of nitrogen to the watershed of the Chesapeake Bay. Nitrogen loading from the nitrate component of deposition (the part of atmospheric deposition caused by nitrogen oxide emissions) is approximately equal to that from fertilizers, and is not far short of that from animal wastes.



F. Estimated Nitrogen Loadings to the Bay

We use two different methods to estimate, from the watershed loadings listed in Table I, the annual average nitrogen loading to the Bay from each source sector. As a "first cut" estimate, we make the approximation that the contribution of each non-point source (atmospheric deposition, fertilizers, and animal wastes) to NPS runoff into the Bay is proportional to the contribution of that NPS to loading of the entire watershed. If a proportional amount of each non-point source reaches the Bay, we expect 24.9% of the NPS runoff reaching the Bay to originate from atmospheric nitrate deposition. This represents 16.6 million kg nitrogen/year in a year of average rainfall, based on the EPA Chesapeake Bay Program estimate of total NPS contribution to nitrogen loading. (cf. Table IV)

The EPA estimate of nitrogen loading does not include the contribution of atmospheric deposition falling directly on the Bay itself. Since the Bay occupies 1.14 million hectares of the area of the watershed, we can use the nitrate and ammonium deposition rates at the nearest NADP monitor (Wye, See Table II) to calculate that 8.3 million kg/year of nitrate nitrogen and 4.6 million kg/year of ammonium nitrogen will be deposited directly into the Bay. The total from direct atmospheric nitrate deposition to the Bay plus the atmospheric nitrate component of land runoff would then be 24.9 million kg/year in

Table IV

## FIRST ESTIMATE OF CONTRIBUTION OF NITROGEN SOURCES TO CHESAPEAKE BAY\*

|                                       |                      |
|---------------------------------------|----------------------|
| Point Source Contribution             | 32.9 million kg N/yr |
| Non Point Sources                     |                      |
| atmospheric nitrate                   | 16.6                 |
| atmospheric ammonium                  | 9.2                  |
| fertilizer                            | 18.4                 |
| animal waste                          | 22.6                 |
| NPS Contribution                      | 66.8                 |
| Subtotal, excluding direct deposition | 99.7                 |
| Deposition to surface of Bay:         |                      |
| atmospheric nitrate                   | 8.3                  |
| atmospheric ammonium                  | 4.6                  |
| Subtotal, direct deposition           | 12.9                 |
| TOTAL                                 | 112.6                |

Table IV-B

## FIRST ESTIMATE OF PERCENT CONTRIBUTION OF NITROGEN SOURCES TO THE BAY

|                      |       |
|----------------------|-------|
| Point sources        | 29.2% |
| Atmospheric nitrate  | 22.1  |
| Atmospheric ammonium | 12.3  |
| Fertilizer           | 16.3  |
| Animal waste         | 20.1  |

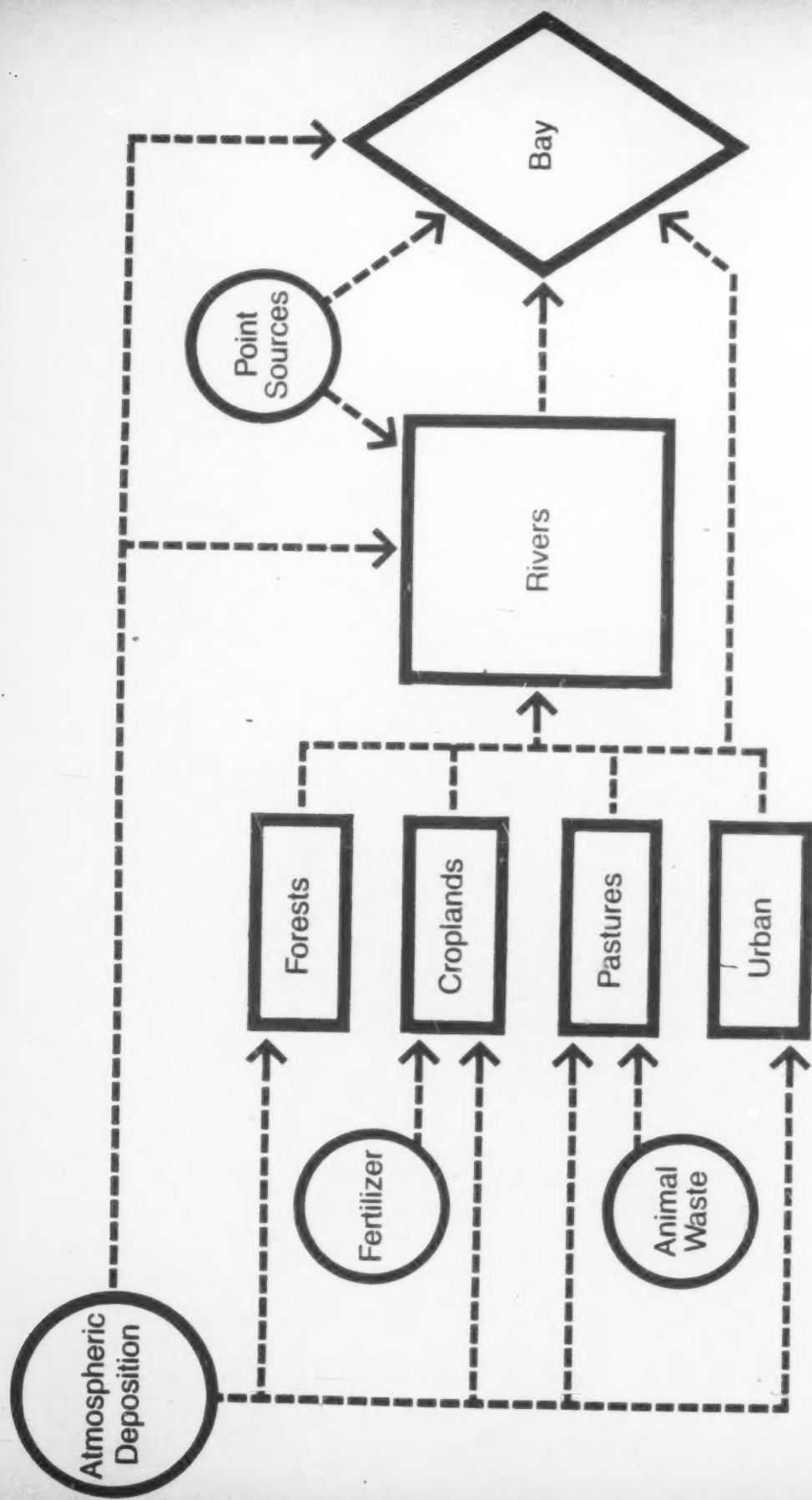
\*Totals for point source contribution, and NPS contribution are from reference 52. Estimated breakdown of NPS by category as described in Discussion and Results. Reference 52 does not include the contribution of direct deposition to the surface of the Bay, so we modify the total by this amount.

an average rainfall year. This represents 22% of the calculated total nitrogen loading to the Bay.

However, it is possible that some of the non-point sources we have discussed will be retained in terrestrial or riverine systems more or less efficiently than the others, causing some to contribute a disproportionately larger or smaller share of the total NPS loading to the Bay than that which would be predicted based solely on total watershed loadings of each.

For instance, since much of the nitrogen in animal waste is associated with particulate organic matter, one might expect this form of nitrogen to be retained more efficiently by the watershed than the more soluble nitrogen in fertilizers or atmospheric deposition. To improve our estimate for the atmospheric deposition contribution to the Bay loadings, we have calculated separately its contribution to the nitrogen loadings on the various land types found in the watershed. We then estimate, for each land type, what fraction of input nitrogen is expected to reach the Bay. This procedure is shown schematically in Figure 4. The nitrogen inputs that we calculate for each of the land uses are summarized in Table V. The loadings we estimate to the Bay from this analysis are shown in Table VI-A and VI-B.

Atmospheric nitrogen deposition and point source nitrogen are the only sources which are input directly into the Bay itself. A total of 26.6 million kg is estimated by the Bay Program to be discharged by point sources directly into the



Nitrogen Flow in the Watershed

Figure 4

Table V  
ANTHROPOGENIC NITROGEN INPUTS TO THE WATERSHED, BY LAND USE

|                      |      |                          |
|----------------------|------|--------------------------|
| Direct Bay Inputs    |      |                          |
| Point Sources        |      | 26.6 million kg nitrogen |
| Atmospheric Nitrate  |      | 8.3                      |
| Atmospheric Ammonium |      | 4.6                      |
| Forests              |      |                          |
| Atmospheric Nitrate  |      | 89.5                     |
| Atmospheric Ammonium |      | 49.5                     |
| Croplands            |      |                          |
| Fertilizer           | 158  |                          |
| Atmospheric nitrate  | 25.6 |                          |
| Atmospheric ammonium | 14.1 |                          |
| Pastures             |      |                          |
| Animal waste         | 195  |                          |
| Atmospheric nitrate  | 23.6 |                          |
| Atmospheric ammonium | 13.0 |                          |
| Urban                |      |                          |
| Atmospheric nitrate  | 4.3  |                          |
| Atmospheric ammonium | 2.4  |                          |



Table VI-A

REFINED NITROGEN LOADINGS TO THE BAY, BY LAND USE  
(Millions of Kilograms of Nitrogen Per Year)

|                            |      |       |
|----------------------------|------|-------|
| Point Sources*             |      |       |
| Below Fall Line            | 26.6 |       |
| Above Fall Line, delivered | 6.3  |       |
| Subtotal                   |      | 32.9  |
| Direct Deposition          |      |       |
| Atmospheric Nitrate        | 8.3  |       |
| Ammonium                   | 4.6  |       |
| Subtotal                   |      | 12.9  |
| Croplands                  |      |       |
| Fertilizer                 | 47.7 |       |
| Atmospheric Nitrate        | 7.8  |       |
| Atmospheric Ammonium       | 4.3  |       |
| Subtotal**                 |      | 59.8  |
| Forests                    |      |       |
| Atmospheric Nitrate        | 9.0  |       |
| Atmospheric Ammonium       | 5.0  |       |
| Subtotal                   |      | 14.0  |
| Pastures                   |      |       |
| Animal Waste               | 4.9  |       |
| Atmospheric Nitrate        | 7.1  |       |
| Atmospheric Ammonium       | 3.9  |       |
| Subtotal                   |      | 15.9  |
| Urban                      |      |       |
| Atmospheric Nitrate        | 2.8  |       |
| Atmospheric Ammonium       | 1.6  |       |
| Subtotal                   |      | 4.4   |
| TOTAL                      |      | 139.9 |

\* From Reference 52

\*\* Cropland total from Reference 52, estimated contribution of individual sources to this total as described in Discussion and Results

Table VI-B  
REFINED ESTIMATE OF NITROGEN BY INPUT SOURCE

| Source               | Total, million kg | % contribution* |
|----------------------|-------------------|-----------------|
| Point Sources        | 32.9              | 24%             |
| Atmospheric Nitrate  | 35.0              | 25%             |
| Atmospheric Ammonium | 19.4              | 14%             |
| Fertilizer           | 47.7              | 34%             |
| Animal Waste         | 4.9               | 4%              |

\* Does not total to 100% due to independent rounding

Bay<sup>52</sup>. As discussed above, we calculate that 8.3 million kg of nitrate nitrogen and 4.6 million kg of ammonium nitrogen reaches the Bay by falling directly into it. Thus the total amount of direct nitrogen input to the Bay itself is 39.5 million kg per year, of which 21% is nitrate deposition to the Bay's surface. Clearly, atmospheric deposition is a significant source of direct nitrogen input to the Bay. Neither of the other nitrogen sources we consider here, fertilizers or animal wastes, is input directly to the Bay, but reaches it via land runoff.

Another 6.3 million kg of nitrogen per year is estimated by the Bay Program to reach the Fall Line from point sources located above it in a year of average rainfall. If we compare this value to the Bay Program's estimate for total "end of the pipe" nitrogen discharged above the Fall Line (12.6 million kg<sup>12</sup>) we calculate that nitrogen loads are reduced 50% by river uptake before the nitrogen reaches the Bay. This retention factor is expected to change in the revised model<sup>12</sup>, but for our purposes it should suffice as an estimate of load reduction due to river uptake.

The rest of the nitrogen inputs to the watershed are land inputs, of which some fraction reaches the Bay via land runoff. Of course, how much of the nitrogen runs off the land depends on the kind of land, and the different types of nitrogen inputs to it.

For each land use shown in Figure 4, it is possible to

estimate the amount of nitrogen input from atmospheric deposition and compare it to nitrogen input from other sources. We find that for all land uses, atmospheric deposition represents a significant source of anthropogenic nitrogen, and in some cases it is the predominant source (See Table V). We then estimate how much nitrogen will run off each land type. We base our estimates on existing studies of nitrogen retention by land type. Such studies are limited in both number and scope, and generally do not examine watersheds as large and complex as that of the Chesapeake Bay. Further studies should be done to improve our understanding of how nitrogen is cycled, especially by large watersheds. The role of atmospheric deposition in this cycles merits particular attention. However, the values below represent best estimates based on current understanding.

The Bay Program estimates that 17.9% of the land in the Chesapeake Bay watershed is cropland (about 2.93 million hectares).<sup>8</sup> If we assume that the entire 158 million kg/ yr of fertilizer nitrogen input we have calculated for the watershed is applied uniformly to the total cropland, we obtain a fertilizer application rate of 54 kg nitrogen/ha-yr. We calculate that the average deposition rate of atmospheric nitrate to the watershed is 8.71 kg  $\text{NO}_3\text{-N}$ /ha-yr, and for ammonium the deposition rate is 4.81 kg  $\text{NH}_4\text{-N}$ /ha-yr, giving total inputs to cropland of 25.6 million kg from nitrate deposition and 14.1 million kg from ammonium. If the main inputs to croplands are fertilizer and atmospheric deposition,

we calculate total nitrogen inputs to be 67.4 kg N/ha-yr, of which 13% is from atmospheric nitrate deposition.

Since both fertilizer and atmospheric deposition contain primarily soluble, inorganic forms of nitrogen, we don't expect the cropland ecosystem to process fertilizer nitrogen in a substantially different way than atmospheric nitrogen. It should therefore be a good approximation to assume that since atmospheric nitrate deposition represents about 13% of the loading to croplands, about 13% of the nitrogen in runoff from croplands can be attributed to atmospheric nitrate.

The Bay Program estimates that in a year of average rainfall, 60%, or 59.8 million kg nitrogen/yr of the total nitrogen loading to the Bay originates from croplands. For croplands, the Bay Program runoff estimates are based on total nutrient loads observed to leach from this type of land - (using wash off of soil as a surrogate for nutrient leaching). The model does not explicitly examine nitrogen runoff as a function of nitrogen inputs. However, since the measured runoff will include any nitrogen derived from atmospheric deposition, for this land type we believe it appropriate to use the calculated delivered load, and estimate the fraction from atmospheric deposition. If we assume outputs are proportional to inputs, the cropland runoff of 59.8 million kg nitrogen/yr can be broken down into 7.8 million kg nitrogen/yr of nitrogen from atmospheric nitrate deposition, 4.3 million kg from atmospheric ammonium, and 47.7 from fertilizer. If we compare



total loadings to croplands (197.7 million kg nitrogen) to the predicted delivered load to the Bay, we calculate an overall nitrogen retention rate from croplands in the entire Bay watershed of 70%, including river uptake.

The Bay Program estimates that all other land uses combined contribute only 7 million kg of nitrogen per year to the Bay (average rainfall), an estimate which seems quite low considering that these uses occupy 82.1% of the watershed and that the nitrogen inputs to them are considerable. For land uses other than cropland, the Bay Program watershed model derives its estimates by calculating nitrogen loadings to each land type between storm events. Since nitrogen deposition is not included in nitrogen loadings to these land types, it is not included in the runoff, which may explain the low estimate. For these land types, we have therefore developed separate estimates of delivered loads to the Bay, based on retention factors from various watershed studies.

The bulk of the watershed (62.6%) is forested. The main source of external nitrogen to forests is atmospheric deposition. Thus, most of the nitrogen in runoff from forests can be attributed to atmospheric sources. Although nitrogen runoff per hectare is lower for forests than for other land uses, this land use occupies such a large fraction of the watershed that total loadings of nitrogen from forested areas to the Bay could still be quite large.

Many studies have been conducted to determine the amount of

nitrogen from atmospheric deposition which is retained by forested watersheds.<sup>46,54,56,58</sup> The results show that anywhere from 50-97% of the nitrogen is retained, depending on the specific features of the watershed. Thus, forested lands represent a potential source of 2,690,000 to 45,800,000 kg per year of nitrogen which ultimately comes from atmospheric nitrate. The study in which 97% retention was observed<sup>56</sup> (based on bulk precipitation vs. stream output) was conducted in a coastal plain watershed. The high retention is probably due in part to the flatness of the landscape, since steeper slopes cause higher runoff. Most of the forested land in the Chesapeake watershed is located in the hillier Piedmont regions above the Fall Line, so the fraction of nitrogen retained is expected to be less. In a study of 3 forested watersheds in Tennessee<sup>46</sup>, best estimates for retention were 66 to 79% of total deposition.

For our purposes, we have chosen 80% as a reasonable mid-range estimate of the amount of nitrogen retained by forested systems. Using this value, we calculate that 17.9 million kg of nitrogen per year will run off forested lands. This represents the amount we expect to reach streams. For streams above the Fall Line, some additional uptake of nitrogen may occur before the loads reach the Fall Line. The Bay watershed model predicts an additional 50% reduction of river loads before they reach the Fall Line. We make the conservative estimate that all forest runoff will be decreased by this

amount. This amounts to assuming that all of the forested lands are located above the Fall Line, which yields a conservative estimate because those forests located below the Fall Line will not have their runoff reduced by river uptake before reaching the Bay. In this manner, we arrive at a final estimate of 9.0 million kg of nitrogen per year reaching the Bay as runoff of atmospheric nitrate deposition and 5.0 million kg from atmospheric ammonium from forested land.

Nitrogen retained in forests is not permanently removed from the watershed drainage system. A disturbance, such as insect invasion, clear-cutting, or climatic change can lead to discharge of large quantities of nitrogen to surface waters.

Another 16.5% of the land in the watershed is estimated to be pastures.<sup>8</sup> This is the main land type which will be receiving animal waste inputs. If we assume that animal waste is input uniformly across this land type, the input rate is 72 kg/ ha-yr. Pastures will also receive 23.6 million kg in atmospheric nitrate input, and 13.0 million kg from atmospheric ammonium.

It has been estimated that only about 5% of animal waste applied to land reaches streams via runoff.<sup>11</sup> This low transfer to streams is presumably due to the fact that a significant amount of nitrogen in animal waste is associated with particulate organic matter, and is thus readily retained. Some animal waste nitrogen is also volatilized to as ammonia. On the other hand, the soluble nitrogen in rainfall will not be

so readily retained. To calculate pastureland contribution to nitrogen loadings of the Bay, we assume that 95% of the nitrogen in animal waste will be retained, and that this will be further reduced by 50% due to river uptake. To estimate the retention of the more soluble nitrogen in rainfall, we assume that the average retention factor will be similar to that of croplands (70%, including river uptake). We expect the distribution of pastures throughout the watershed to be similar to the distribution of croplands, since both are located in agricultural regions. Using these assumptions, we estimate that pasturelands contribute 4.9 million kg nitrogen per year from animal waste, 7.1 million kg from atmospheric nitrate, and 3.9 million from ammonium deposition.

The rest of the watershed (3%) is urban land. Although this land use occupies only a small area of the watershed, we expect its contribution to runoff to be disproportionately large. All of the major cities (Baltimore, Washington, Richmond, and the Norfolk-Hampton complex) are located at or below the Fall Line, so any runoff from these areas goes directly into the Bay. Urban areas are largely covered by impervious surfaces, such as asphalt and concrete, which will not retain any nitrogen at all. For these reasons, we expect the retention factor for this land use to be lower than for any other land use. We therefore expect urban land to retain somewhere between 70% (same as croplands) and 0% (complete washoff of all nitrogen) of nitrogen inputs. We chose 35%



retention as a mid-range estimate. In this manner, we calculate that urban lands will contribute 2.8 million kg of atmospheric nitrogen, and 1.6 million kg of atmospheric ammonium.

Although we have performed an analysis based on annual average values, it is important to recognize that seasonal variations do occur, which can alter the relative importance of each source sector in any given season. For instance, fertilizer input is disproportionately important in the spring, because most fertilizer is applied during this season. Nitrogen runoff from forests is also higher in early spring, especially in northern portions of the watershed, due to nitrogen input from snowpack melt, and decreased nitrogen retention due to frozen ground and absence of growing plants. Direct input of nitrogen to the Bay itself, both from point sources and from atmospheric deposition, becomes disproportionately important in the summer because river flows are at their lowest during this season. Our estimates serve to identify the relative significance of the various source sectors to annual average loadings to the Bay. Further analyses would be needed to provide insights which are important for designing nutrient control strategies which take into account seasonal differences.



## G. Discussion of Results

Summing contributions from all land uses, we calculate that the total external nitrogen loading to the Bay is 139.9 million kg of nitrogen per year. Although this is higher than the Bay Program's estimate for an average rainfall year, it falls within the range defined by dry and wet years (84.0 to 179.5 million kg nitrogen per year). It is also in reasonable agreement with the sum of USGS measured total river inputs of the three biggest tributaries (average for 1979 and 1980 is 135.5 million kg nitrogen per year, see page 38 in reference 48) plus point source inputs below the Fall Line and atmospheric deposition on the Bay, which sums to 174.7 million kg nitrogen per year. The 135.5 total for river inputs is based on only two years of monitoring and 1979 was a high flow year. Based on more representative flow conditions estimated river input is 89.1 million kg of nitrogen per year (see p. 231, Reference 38), and total average inputs of external nitrogen to the Bay are 123 million kg of nitrogen per year.

Our refined estimate of the contribution of atmospheric nitrate deposition to the total loading of the Bay is 25% (See Figure 5). This is somewhat higher than our estimate by our first method, in which we did not take into account the relative higher solubility of precipitation nitrogen compared to animal waste. Our revised estimate of fertilizer loading is 34%,

## Sources of Nitrogen—The Bay

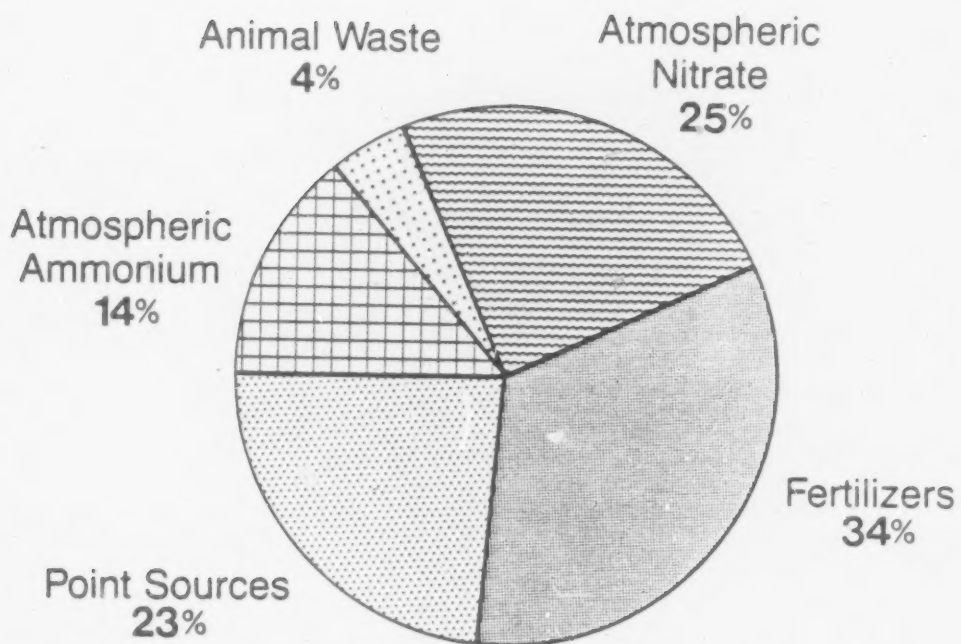


Figure 5

which takes into account the high rate of runoff from croplands. We estimate that animal waste only contributes 4% to the loadings. This is probably a low estimate, since we have based it on a high retention factor (95%). Our retention factor assumes that all of the animal waste is land applied, however other manure management practices, such as storing liquid waste in lagoons (which occasionally rupture), or allowing direct discharge to waterways may increase the amount of animal waste nitrogen reaching the Bay<sup>10</sup>. A significant fraction of the nitrogen input from atmospheric ammonium also originates from animal waste<sup>15</sup>

From the above analysis, it is clear that atmospheric nitrate represents a significant source of nitrogen to the Bay. In addition to sizable inputs from deposition directly to the Bay itself, we expect a significant fraction of land runoff to ultimately derive from atmospheric sources. We consider 25% to be a conservative estimate of the amount of nitrogen loading to the Bay which ultimately comes from atmospheric nitrate deposition. For instance, if nitrogen inputs to forested land are only reduced by 66% before reaching rivers, rather than 80%, the total atmospheric nitrate contribution climbs to 29% of the total loading to the Bay.

#### H. Uncertainties

The error in the wet component of the atmospheric contribution due to imprecision in sampling and analysis by the NADP laboratories is less than 10 percent.<sup>59</sup> The other sources of uncertainty in calculation of the atmospheric contribution are discussed above (amount of dry deposition, differences in deposition at high and low elevations). Uncertainties in the other external source values (fertilizers, animal waste, and point sources) are difficult to estimate, and quantitative error analyses are generally lacking in the published reports from which we obtained this data. In the case of the contributions to the Bay itself, the uncertainty in watershed and riverine retention probably dominates all others. We have provided a range of estimates in the preceding discussion for that source of uncertainty.

## I. Future Nitrogen Loadings to the Bay

In the preceding analysis, we calculated the contribution of atmospheric nitrate deposition to current nitrogen loadings of the Bay. In this section we estimate future loadings, given the current strategies for controlling nutrient enrichment of the Bay.

The Chesapeake Bay agreement<sup>60</sup> sets a goal of 40% reduction of nitrogen loading to the Bay. To calculate future loadings, we assume that this goal will be achieved for those nitrogen source sectors currently being considered for reductions, point sources and agricultural runoff. Most atmospheric ammonium originates from agricultural sources, primarily from decomposition of animal waste, with volatilization of fertilizer also contributing<sup>15</sup>. It is not clear whether agricultural best management practices (BMP's) designed to reduce runoff will also reduce the agricultural contribution to atmospheric ammonium. We decided to make the optimistic assumption that BMP's would reduce atmospheric ammonium by 20%. In the absence of further nitrogen oxide emission controls, emissions in the regions which most strongly influence deposition to the Chesapeake Bay region are predicted to increase by 44% by the year 2030<sup>14</sup> (See Section J below). We assume that atmospheric nitrate deposition to the Bay's watershed, and nitrogen loading to the Bay from this source will increase proportionately to nitrogen oxide emissions.



In Table VII we list our calculated future loadings to the Bay. Figure 6 shows the fraction of nitrogen loading that each source is expected to contribute in the year 2030. In the absence of nitrogen oxide controls, the contribution of atmospheric nitrate deposition to nitrogen loading of the Bay will climb to 42% of the total, becoming the major source sector. Instead of achieving the desired 40% reduction of nitrogen in the Bay, we calculate that the reduction will only be 16%.

Table VII  
ESTIMATED FUTURE NITROGEN LOADINGS TO THE CHESAPEAKE BAY

| Source               | Current | Future (2030) |     |
|----------------------|---------|---------------|-----|
| Atmospheric Nitrate  | 35.0    | 50.4          | 42% |
| Atmospheric Ammonium | 19.4    | 15.5          | 13% |
| Point Sources        | 32.9    | 19.7          | 17% |
| Fertilizer           | 47.7    | 28.6          | 25% |
| Animal Waste         | 4.9     | 2.9           | 3%  |
| TOTAL                | 139.9   | 117.1         |     |

## Future Nitrogen Loading to The Bay

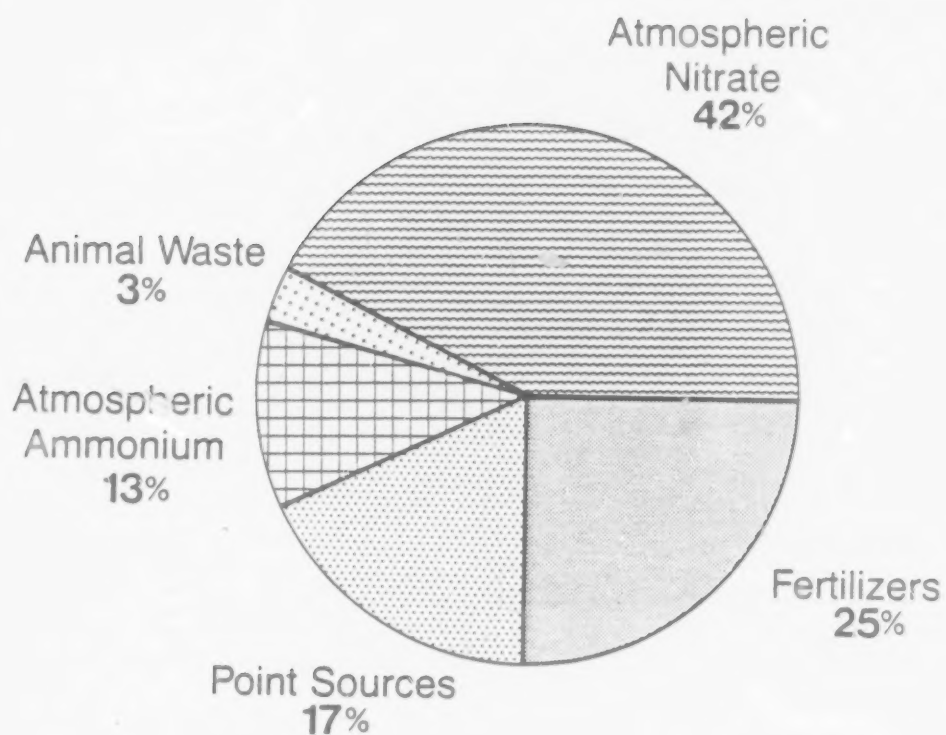
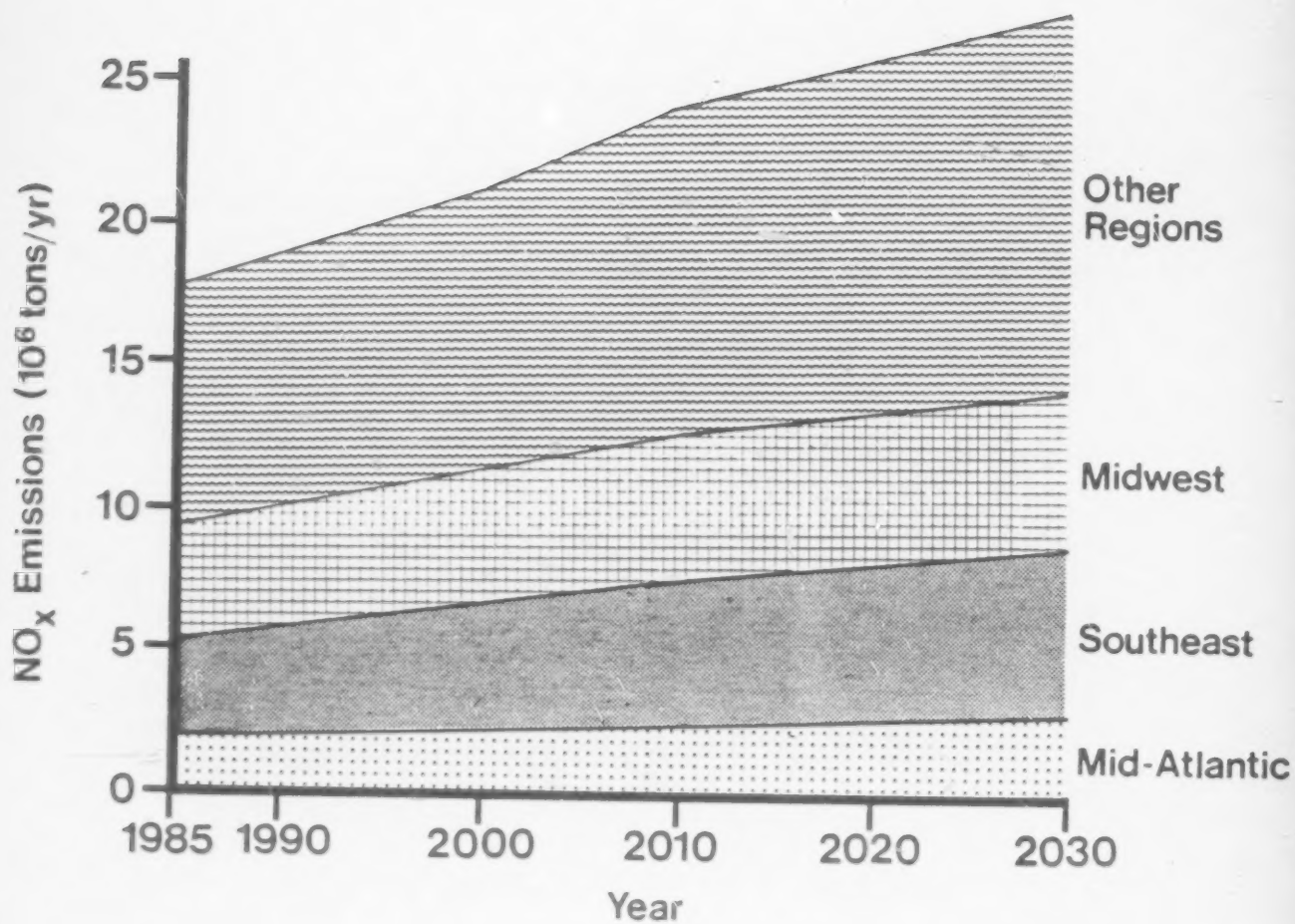


Figure 6

J. Sources of Atmospheric Nitrate Deposition to the Watershed

Much of the nitrate which is deposited to the Chesapeake Bay's watershed originates from sources located hundreds of miles upwind. For instance, in the state of Maryland, an estimated 70-90% of nitrate deposition originates from outside the state.<sup>61</sup> Storm trajectory analysis indicates that most of this nitrate originates in areas to the west, and southwest of Maryland.<sup>62</sup> In New York, 80-85% of the nitrate deposition originates outside the state<sup>63</sup>. Similar patterns are expected to hold for other states in the watershed. The two most important sources of nitrogen oxides emissions in the U.S. are highway vehicles, which contribute 36% of the total, and power plants, which contribute 32%<sup>13</sup>. Other transportation activity contributes an additional 6 to 10%<sup>14</sup>.

In the absence of any additional controls, emissions of nitrogen oxides are expected to increase substantially over the next 40 years, in all regions of the U.S., including those regions whose emissions influence nitrate deposition rates to the Chesapeake watershed (See Figure 7, adapted from Reference 14). In the Mid-Atlantic region, which contains all the states in the Chesapeake's watershed except New York, nitrogen oxide emissions are projected to increase 38% by the year 2030. In the midwestern region, to the west of the watershed, emissions are expected to increase 33%, and in the region to the southwest, the expected increase is 63%<sup>14</sup>. The combined



NO<sub>x</sub> Emissions are Projected to Increase

Figure 7



increase for these 3 regions by 2030 is 44%.

In the absence of further controls on nitrogen oxide emissions, it is clear that nitrogen loading to the Chesapeake Bay from atmospheric sources will increase substantially over the next forty years. If strategies are pursued to reduce other sources by 40%, but not atmospheric nitrate deposition, we estimate that the contribution of this source will climb to 42% of the total loading nitrogen loading of the Bay. Instead of achieving the goal of 40% reduction, nitrogen loading will decrease by no more than 17%. Potential improvements in water quality of the Bay due to decreased nitrogen inputs from other sources will be cancelled by increases from atmospheric sources.

K. Recommendations for the Chesapeake Bay Watershed

In order to improve water quality in the Chesapeake Bay, large reductions of nitrogen inputs will be necessary. In fact, the U.S. government and the governments of Virginia, Pennsylvania, Maryland and Washington D.C. recently agreed to goals of 40% reduction of inputs of nitrogen and phosphorus to the Chesapeake Bay<sup>60</sup>. Since there are practical limits to the amount any one source can be reduced, it makes sense, whenever possible, to consider reductions in all source sectors. Programs are being considered to reduce runoff from agricultural sources, and control of point source nitrogen has also been proposed<sup>64</sup>. We support these proposals. However, the contribution of nitrogen from atmospheric nitrate deposition must be taken into account, since this is also a major source. In the absence of controls, this source is expected to increase by approximately 40% in the next forty years. Technologies are readily available for controlling nitrogen oxides emissions both from motor vehicles and power plants. Implementation of such technologies should be included in any strategy for nutrient reduction to the Bay.

There are some measures which can be taken at the state level to reduce nitrogen oxide emissions. These measures include incentives for energy efficiency in electricity use, improved inspection and maintenance of vehicles to assure that current emission standards for nitrogen oxides are being met,

and more stringent emission standards for nitrogen oxides from power plants and industrial sources. Such measures should be enacted in all of the states within the Chesapeake Bay watershed.

However, there are limits to what state level nitrogen oxide control programs can accomplish. This is due both to the large amounts of nitrogen oxides transported into the region from upwind sources, and federal regulations prohibiting states from establishing state standards for nitrogen oxide emissions from vehicles.

The states of the Chesapeake Bay watershed are currently planning to pursue strategies to reduce nitrogen inputs to the Bay from agricultural runoff, and point sources. Unless nitrogen oxide emissions are also controlled, much of the reductions in nitrogen from these other sources will be undone by increases from atmospheric deposition. Federal legislation mandating more stringent emission standards both for automobiles and for power plants and other stationary sources is needed. Legislators from states within the Chesapeake Bay region should aggressively pursue such legislation. Such an approach will safeguard any improvements in water quality of the Bay that these states achieve, and will help one of our nation's most valuable resources, the Chesapeake Bay, on the long road to recovery.

We recommend a decrease in atmospheric deposition to the Bay of 40%, consistent with the planned reductions in other

sources. To achieve such a reduction, emissions from all nitrogen oxide sources affecting the Bay should be reduced accordingly.

## VII. Nitrogen in Other Coastal Waters and Estuaries

A. New York Bight: In the summer of 1976, coastal waters of New Jersey suffered from a severe depletion in dissolved oxygen, leading to large fishkills, including a 60% reduction in the clam harvest for that year. The immediate losses were estimated at \$62 million, and cumulative losses in later years due to lost stock were estimated to be as high as \$550 million. Although the causes of this episode are not completely understood, nutrient enrichment was a contributing factor. Indeed, nutrient enrichment has contributed to low oxygen levels in the Bight every year since 1976<sup>65</sup>.

In order to better understand this episode, NOAA conducted a study which included a nutrient budget for the Bight. NOAA estimated that atmospheric deposition contributed 13% to the overall annual nitrogen loading to the New York Bight<sup>66</sup> (separate estimates for contribution of nitrate and of ammonium were not available). Like most nitrogen budgets to coastal waters, this study only estimates the contribution of atmospheric deposition via direct precipitation falling on the Bight itself. Another 29% of the nitrogen input comes from runoff. A significant fraction of the nitrogen in runoff could also originate in atmospheric deposition to land and rivers. Atmospheric nitrate deposition appears to be an important contributor to the problems of this nutrient-enriched body of water.



B. Long Island Sound: NOAA <sup>67</sup> has estimated total nitrogen loadings to the Sound from point sources, and land runoff at 45.5 million kg of nitrogen per year. This estimate did not include input from atmospheric deposition. Our preliminary estimates, assuming a deposition rate of 9.34 kg  $\text{NO}_3\text{-N/ha-yr}$  to the Sound and its drainage area (wet deposition at West Point, the nearest NADP monitor, was 4.67 kg/ha-yr in 1984, we assume dry deposition is approximately equal), indicate that about 41.7 million kg/yr of nitrogen from nitrate deposition falls on the land that drains into the Sound, contributing to runoff. About 3.1 million kg/yr of atmospheric nitrate deposition, or about 7% of the total external nitrogen budget, falls directly into the Sound itself.

In our Chesapeake Bay analysis, we estimate a total loading to the Bay from atmospheric nitrate of 35 million kg of nitrogen per year, including 8.3 million from direct deposition, and another 26.7 million from land runoff. About 19% of total atmospheric nitrate deposition to the watershed is delivered to the Bay. If we assume the same fraction is delivered from land to the Sound, we estimate 7.9 million kg of nitrogen/yr reaching the Sound from land, and 3.1 by direct deposition to water, for a total of 11.0 million kg of nitrogen. Thus, 23% of the total nitrogen load to the Sound originates in atmospheric deposition.

Although we expect point source inputs to play a larger role in river inputs in the Sound's nitrogen budget than in the

Chesapeake Bay's budget, we expect on the other hand that agricultural sources will play a much smaller role. The preliminary values we present for the Sound suggest that atmospheric nitrate is an important part of the external nitrogen budget of the Sound, as it is for the Chesapeake Bay, thus contributing to the eutrophication problems of the Sound.

C. Other Coastal Waters The problem of enhanced primary productivity from atmospheric nitrogen inputs appears to be widespread along the Atlantic seaboard. The Delaware Bay is extremely enriched in nitrogen, especially nitrate nitrogen. Concentrations of nitrate in this bay are 15 times higher than those observed in pristine areas, making the Delaware Bay one of the most nitrate enriched estuaries in the country<sup>68</sup>. This bay has not experienced the same explosive algal blooms observed in some other nutrient enriched waters, probably because high flows of water through this bay do not give blooms an opportunity to form. However, high levels of chlorophyll a (a measure of phytoplankton concentration) and low levels of dissolved oxygen (3 mg/liter) are frequently observed in the spring<sup>68</sup>.

The Albemarle-Pamlico Sound, a large and productive estuary which dominates the coast of North Carolina, has also been plagued with nutrient enrichment, nuisance algal blooms, low oxygen episodes, and frequent fishkills<sup>69</sup>. Harmful algal blooms have been especially severe in two sub-estuaries, the

lower river basins of the Chowan<sup>70</sup> and the Neuse Rivers<sup>71</sup>. As discussed in Section VI above, Paerl has shown that the amount of nitrogen input to the Neuse River is a controlling factor in severity of summer algal blooms<sup>41</sup>.

The North Carolina Division of Environmental Management has calculated that 18% of the nitrogen input to the lower Neuse comes from precipitation nitrogen falling directly on the water<sup>71</sup>. The total nitrogen loading to this estuary is 7.5 million kg per year, including precipitation nitrogen<sup>71</sup>. Using the 1986 nitrate nitrogen deposition rate at the nearest NADP monitor and doubling it to account for dry deposition, we calculate 0.53 million kg nitrogen/yr of nitrate nitrogen falls directly on the water, and another 6.1 million kg falls on land. If we assume that 19% of the nitrate deposition to land reaches the water (as in the Chesapeake basin), the total nitrogen load to the lower Neuse due to nitrate deposition is 1.7 million kg nitrogen/year, or 23% of the total.

Paerl has also documented increases in primary productivity in coastal waters off the North Carolina coast due to inputs of nitrogen from precipitation<sup>24</sup>.

Nitrogen enrichment is a problem in some European coastal waters as well. Both the Baltic and the North Seas have had increasing problems with nuisance algal blooms, which have been most severe in the most nitrogen enriched areas.<sup>72,73</sup> In the Baltic Sea, atmospheric nitrogen input (wet and dry deposition of nitrate and ammonium) from direct precipitation has been

estimated to supply 25% of external nitrogen to the sea.<sup>72</sup>

### VIII. Summary and Recommendations

In summary, we conclude that increases in emissions of nitrogen oxides to the atmosphere over the United States, have contributed to elevated inputs of nitrate to estuaries and other surface waters. Atmospheric nitrate deposition is a major cause of eutrophication problems in estuarine and other coastal systems where nitrogen is a limiting nutrient. The importance of atmospheric nitrate will increase in the future if nitrogen oxide emissions are not further controlled.

Reductions in nitrogen oxide emissions would benefit the environment in many ways. Some of the benefits include:

1. Reduced nutrient flows to coastal waters. Decreased inputs of nitrogen to estuaries and coastal waters will improve the condition of waters which are currently suffering from eutrophication caused by excessive levels of this nutrient.
2. Healthier drinking water. Atmospheric deposition has been shown to be increasing levels of nitrate, a health hazard, in the Catskill reservoir system supplying New York City. Similar trends are expected in most drinking water supplies which rely on surface water.



3. Less acid deposition. Nitric acid is responsible for up to 40% of the acidity of rainfall in the eastern U.S. It also plays a disproportionate role in "acid shock" effects during spring snowmelt.
4. Less formation of ground level ozone (a major component in urban smog). Nitrogen oxides are important precursors to ozone, and may control regional ozone levels over the eastern U.S.

For the U.S., year 2010 nitrogen oxide emissions are expected to increase over 1980 levels by about 30 percent, and by 2030 emissions are forecast to increase by 49 percent<sup>14</sup>. In those regions most influencing nitrate deposition to the Chesapeake Bay, (Mid-Atlantic, Southeast, and Midwest) nitrogen oxide emissions are projected to increase 44% by 2030<sup>14</sup>. In light of the potential deleterious effects on human health and aquatic ecosystems, measures should begin now to decrease these anthropogenic inputs. Controls on nitrogen oxides should be included in a national plan to control the precursor emissions to acid deposition.

Nitrogen flow reductions proposed to protect Chesapeake Bay may be used as a guide to the level of nitrogen emissions reduction in the eastern U.S. The Chesapeake Bay plan has a target reduction of 40% for other important nitrogen source sectors.<sup>60</sup> We propose this target for the atmospheric

deposition (and emissions) sector as well.

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## Appendix I

### REGIONAL INCREASES OF NITRATE IN PRECIPITATION AND SURFACE WATERS

It appears that regional increases in nitrogen oxide emissions, especially in the eastern United States, are creating widespread increases in nitrate concentrations in surface waters across the east. The following studies give further evidence of this phenomenon.

From 1950 to 1970 the United States Geological Survey (USGS) measured nitrate in six major U.S. rivers. <sup>1</sup> Although western rivers showed little change, in the East, a significant upward trend was found in the Delaware and Ohio Rivers. These changes were not related to sources within the watersheds themselves. During this same time period, nitrogen oxide emissions in the East show steady increases. <sup>2</sup>

A recent study conducted by the USGS on water quality trends in the nation's rivers during the period 1974 to 1981 documents wide-spread increases in nitrate concentrations. <sup>3</sup> Total nitrogen increases at network stations were strongly associated with high levels of atmospheric nitrate deposition, particularly in the Ohio, Mid-Atlantic, Great Lakes and Upper Mississippi Basins. Increasing trends in total nitrate concentrations were extremely frequent and widespread, outnumbering decreases four to one. Increasing trends were most frequent east of the 100th meridian. These nitrate increases



resulted in rises of from 20 to 50 percent in the delivery of nitrate to Atlantic coast estuaries, the Gulf of Mexico, and the Great Lakes. Statistical analysis showed increases in total nitrogen to be strongly associated with several measurements of agricultural activity and also with atmospheric deposition. Total nitrogen trends appear more related to non-point than to point sources. The study concluded that atmospheric deposition, in particular, may have played a large role in total nitrogen increases in midwestern and eastern basins.

A study conducted on the nitrogen inputs to Lake Mendota, Wisconsin, a moderately eutrophic lake located in a basin of fertile crop and pasture land, found that precipitation on the surface of the lake contributed more nitrogen than did urban and rural runoff combined. <sup>4</sup> Since precipitation on the watershed itself would be factored into runoff, the contribution from precipitation is actually greater than it appears. Despite the importance of agriculture at this site, precipitation plays the major role as a contributor of nitrogen.

At Hubbard Brook, New Hampshire, annual weighted nitrate concentrations in precipitation in the mid-1970's were 2.3 fold greater than they were in the mid-1960's. The increase in nitrate concentration in precipitation in this remote forested area is attributed to increases in amounts and distribution of atmospheric pollutants.<sup>5</sup> Within the Hubbard Brook watershed, in Mirror Lake, most of the input of nitrate comes from precipitation falling directly on the surface of the lake.

Data from 1970 through 1976 show the average annual input of nitrogen from precipitation falling directly on the lake was 119 kilograms, and from runoff, 81 kilograms. <sup>6</sup>

Although most research has focused on nitrogen deposition in the east, western basins also receive substantial amounts of atmospheric nitrogen. Nitrate inputs from precipitation in Lake Tahoe, California, are extremely important as the lake is currently undergoing eutrophication, and nitrate is the limiting nutrient.<sup>7</sup> Precipitation contributes 1-2 kg/ha of inorganic nitrogen per year to the lake, and output of inorganic nitrogen from the lake is 10 to 33 percent of these precipitation inputs,<sup>8</sup> not unlike the nitrate budget of the New York City's Catskill reservoirs, or Hubbard Brook. In western basins nitrate deposition contributes as much as ten times the basin yield of nitrate,<sup>9</sup> suggesting that it is a major source of nitrate.

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Appendix II  
METHOD OF NITROGEN LOADING CALCULATION FOR THE CHESAPEAKE BAY  
WATERSHED

We chose to compile nitrogen totals for the year 1984 since this was the most recent year for which relatively complete data was available. When data for 1984 was not available, we used data from the year closest to 1984 for which data was available.

Point source inputs: This value was provided to us by Joseph Macknis of the US EPA Chesapeake Bay Program from their computer database. It is the total for the year 1985. This is not expected to differ much from the total for 1984.

Fertilizer Consumption: For Pennsylvania and Virginia, commercial fertilizer usage figures were computed with the use of county data on total nitrogen found in 1984 state agricultural reports.<sup>1,2</sup>

For counties which are only partially included in the watershed, the land area which lies in the watershed was calculated using an Uchida compensation planimeter on USGS hydrological unit maps. The overall error in using this instrument and land acreage values provided in the census is 1.0%. The percentage of the county within the watershed was calculated. The total amount of nitrogen in fertilizer consumed in the county was multiplied by this percentage to estimate amount of fertilizer nitrogen consumed in the part of the county which lies in the watershed.

For those states which did not have 1984 county estimates for fertilizer consumption, an estimate of fertilizer usage was derived as follows. The 1982 Census of Agriculture<sup>3</sup> figures for number of acres in the county on which fertilizer was used, and the total number of acres in the state on which fertilizer was used, were used to derive the percentage of the state fertilizer total used by that particular county. It was assumed that each county would use the same percentage of the state total of fertilizer in 1984 as they used in 1982. The county percentage was then multiplied by the total 1984 fertilizer usage in the state, obtained from state reports<sup>4,6</sup> to derive an estimate of fertilizer used in that county in 1984. For counties not entirely within the watershed, the adjustment described above was made, to estimate usage within the watershed. Table A-I shows the calculated totals for that portion of each state which is in the watershed.

Atmospheric deposition: For each of the drainage basins listed in Table II, it was assumed that wet deposition of nitrogen would be approximately equal to that observed at the nearest NADP station. The 1984 reported deposition value<sup>7</sup>, in grams of nitrate or ammonium per square meters, was converted to kilograms of nitrate nitrogen or ammonium nitrogen per hectare. For each sub-basin, this value was multiplied by the USGS reported value for total number of hectares in the basin to give an estimate for total wet nitrogen deposition in the basin.



Table A-I

TOTAL FERTILIZER NITROGEN APPLIED TO THE CHESAPEAKE WATERSHED  
(million kg nitrogen per year)

| State         | Amount  |
|---------------|---------|
| New York      | 10.825  |
| Pennsylvania  | 37.863  |
| Virginia      | 41.794  |
| West Virginia | 4.841   |
| Maryland      | 56.567  |
| Delaware      | 6.408   |
| Total         | 158.298 |

To calculate total atmospheric nitrogen deposition (wet + dry), we multiply our wet deposition total by a factor of 2. Because dry deposition is difficult to measure accurately, it is often ignored completely. We believe that this is inappropriate, since dry deposition will contribute significantly to atmospheric nitrogen loading to the watershed. Studies of dry deposition of nitrogen have found total nitrogen deposition to be 2 to 5 times wet deposition.<sup>8-11</sup> Therefore, assuming that total deposition is twice the wet deposition represents a conservative estimate of the total atmospheric loading. Total deposition values derived in this manner are listed in Table II.

Animal waste: For states in which 1984 county data was obtained<sup>2,4,6</sup> (all but New York), the total number of any given farm animal in the county was multiplied by the amount of nitrogen that each individual animal excretes each year<sup>12</sup> (See Table A-II) to give total nitrogen produced by that kind of animal in that county. For those counties only partially in the watershed, the percentage contained in the watershed was calculated in the same manner as described above, and the county nitrogen total was then multiplied by the percentage to give an approximation of animal nitrogen produced in the part of the county in the watershed. The watershed totals for each type of animal, in each state, are presented in Table A-III.

Since county data was not available for New York for 1984, the percent of the state total for each animal which was

Table A-II  
NITROGEN PRODUCED IN ANIMAL WASTE\*

| Animal   | lbs Nitrogen/animal-year |
|----------|--------------------------|
| Cattle** |                          |
| Dairy    | 123                      |
| Beef     | 61                       |
| Swine    | 32                       |
| Sheep    | 16                       |
| Poultry  |                          |
| Layers   | 0.94                     |
| Broilers | 0.78                     |
| Turkeys  | 3.04                     |

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\* See Reference 12

\*\* For calves, we assumed that they produce half the nitrogen of full grown cattle in each category of cattle.

Table A-III

## ANIMAL WASTE NITROGEN PRODUCED IN CHESAPEAKE WATERSHED

Millions of kg nitrogen per year\*

| Animal   | NY     | PA     | VA     | MD     | WV      | DE    |
|----------|--------|--------|--------|--------|---------|-------|
| Dairy    | 14.866 | 37.047 | 7.390  | 9.984  | 0.650   | 0.266 |
| Beef     | 1.198  | 6.201  | 17.134 | 3.608  | 2.993** | 0.111 |
| Calves   | 1.046  | 9.877  | 3.498  | 3.476  |         | 0.038 |
| Other    |        |        | 4.334  | 1.440  |         | 0.015 |
| Swine    | 0.186  | 9.135  | 2.756  | 3.111  | 0.301   | 0.353 |
| Sheep    | 0.085  | 0.305  | 0.649  | 0.118  | 0.232   | 0.002 |
| Layers   | 0.612  | 7.913  | 0.957  | 1.854  | 0.152   | 0.158 |
| Broilers | 0.011  | 5.045  | 6.620  | 15.245 | 1.317   | 5.286 |
| Turkeys  | 0.005  | 1.510  | 4.970  |        | 1.051   |       |
| total    | 18.009 | 77.033 | 48.308 | 38.836 | 6.696   | 6.229 |

Grand Total = 195.111

\* totals are for portion of state contained within the watershed

\*\*includes calves, and "other" cattle

Insert Table A-III



contained in each county was derived as described above, using 1982 Census of Agriculture data.<sup>3</sup> This percentage was then applied to the 1984 state totals to estimate number of each kind of animal in each county in 1984.

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